

Observing the evaporation transition in vibro-fluidized granular matter

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Abstract

By shaking a sand box the grains on the top start to jump giving the picture of evaporating a sand bulk, and a gaseous transition starts at the surface granular matter (GM) bed. Moreover the mixture of the grains in the whole bed starts to move in a cooperative way which is far away from a Brownian description. In a previous work we have shown that the key element to describe the statistics of this behavior is the exclusion of volume principle, whereby the system obeys a Fermi configurational approach [1]. Even though the experiment involves an archetypal non-equilibrium system, we succeeded in defining a global temperature, β^{-1} , as the quantity associated to the Lagrange parameter in a maximum entropic statistical description. In fact in order to close our approach we had to generalize the equipartition theorem for dissipative systems. Therefore we postulated, found and measured a fundamental dissipative parameter, δ , written in terms of pumping and gravitational energies, linking the configurational entropy to the collective response for the expansion of the centre of mass (c.m.) of the granular bed. Here we present a kinetic approach to describe the experimental velocity distribution function (VDF) of this non-Maxwellian gas of macroscopic Fermi-like particles (mFp). The evaporation transition occurs mainly by jumping balls governed by the excluded volume principle. Surprisingly in the whole range of low temperatures that we measured this description reveals a lattice-gas, leading to a packing factor, p_f , which is independent of the external parameters. In addition we measure the *mean free path*, L , as a function of the driving frequency, f_e , and corroborate our prediction from the present kinetic theory.

The granular matter bed was set up with 5437 balls of $ZrO_2 - Y_2O_3$, in a glass container of $D_c = 50\text{mm}$ diameter (which is open on the top) in a chamber at 1atm of air under low water vapor content conditions ($5.8 \pm 0.2\text{gr/m}^3$), see Fig. 1(a,b). The diameter of the balls was $D = 1.99\text{mm}$ and their mass $m = 26.8 \pm 0.1\text{mgr}$. Under vertical vibration, it is possible to get the balls at the lower gravitational position of the container in a crystalline state (the bulk), and those at the upper one in a gas state, which is called the fluidized gap [1]. This non-Markovian gas doesn't expand to the whole volume of the container, and the number of the *active* moving particles, in the gas phase, is a function of the pumping energy. Thus L of the mFp determines the occupied volume of the evaporated gas. A sinusoidal vibration is driven by a vibration plate on the GM bed of height h (with intensity $\Gamma = A\omega^2/g$, where A is the amplitude, g is the acceleration of gravity, and $\omega = 2\pi f_e$). The vibration apparatus is set up by an electromagnetic shaker (TIRAVIB 5212) which allows for feedback through a piezoelectric accelerometer for the control of f_e and Γ in the range of 10-7000Hz, and 2 – 40g respectively. The control loop is completed by an Oscillator Lab-works SC121 and a TIRA 19/z amplifier of 1kw. The vertical trajectory (stochastic realization) $z(t)$ of one particle was followed in a window of 12mm with a laser device by using a triangulation method, see Fig. 1(c) and its spectrum is shown in Fig. 1(d). A laser emitter with a spot of $70\mu\text{m}$ and a linear image sensor (CCD-like array) enables a high speed measurement with $100\mu\text{sec}$ sampling. The linear image sensing method measures the peak position values for the light spots and suppresses the perturbation of secondary peaks, which makes possible a

resolution of $1\mu\text{m}$. The shaker and the laser displacement sensor were placed on vibration-isolated tables to decouple them from external perturbations. $z(t)$ were taken with a 9354C Le Croy Oscilloscope of 500MHz. The velocity, $V(t) = dz/dt$, of the mFp was calculated numerically for $\Delta t = 100\mu\text{sec}$ from $z(t)$ registers. The amplitude dispersion $\sigma_z = \sqrt{\langle z(t)^2 \rangle - \langle z(t) \rangle^2}$ and velocity variance $\sigma_V^2 = \langle V(t)^2 \rangle - \langle V(t) \rangle^2$ were obtained from a window of 2 seconds for each pair of registers $\{z(t), V(t)\}$. In Fig. 2(a) we report σ_z against f_e (from 40-100Hz) for fixed $\Gamma = 10$, for a GM bed of $h = 18\text{mm}$. We test experimentally in Fig.2(b) the relation between σ_z and σ_V^2 . Beyond the Gibbs ensembles, it should be possible to conceive a common conceptual framework under the non-equilibrium thermodynamics [2] that involves jammed states, as described by Edwards ensembles [3, 4]. From a granular gas it is possible to get either a jammed state, for example by reducing or quenching Γ at a given f_e [5], or a crystal by slowly scanning up f_e in a fixed accelerated experimental set-up (cooling down). In fact it is possible to see how a non-Brownian gas is condensed into a *fcc* crystal [6].

The non-equilibrium GM temperature is a controversial quantity presenting anisotropy and inhomogeneous characteristics, and its behavior as a function of the external pumping of energy is yet an unsolved question. However we have laid out an experimental set-up that makes it possible to measure the “global” temperature as the Lagrange parameter that appears in [1].

Since energy is constantly being added to the system a nonequilibrium steady state (s.s.) mass profile for the GM bed is reached. We report here, upon measuring the evaporation transition line in a weakly vibrated GM

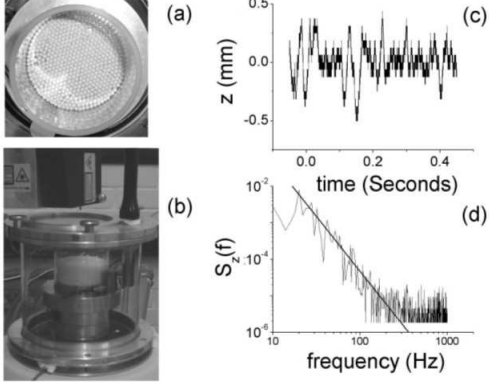


FIG. 1: (a) Top view of the GM bed in a condensed fcc state. (b) Set up for distance measurement (amplitude realizations $z(t)$) corresponding to 11 layer GM ($h = 18\text{mm}$) with $ZrO_2 - Y_2O_3$ balls ($D = 1.99\text{mm}$) in a glass container of $D_c = 50\text{mm}$ diameter. (c) Stochastic realization $z(t)$ at 83Hz, 10g, and (d) the corresponding anomalous power law $f^{-\nu}$ spectrum with $\nu = 3$.

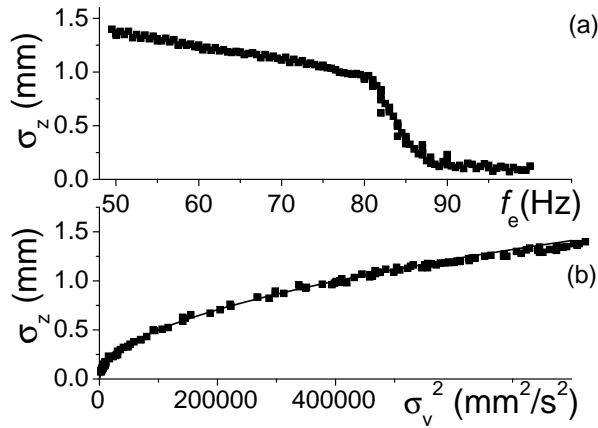


FIG. 2: (a). Evaporation transition for the experiment of Fig.1. σ_z as a function of the external driving frequency f_e . (b) σ_z vs. σ_v^2 and the corresponding theoretical fittings. The f_e was from 50 – 100Hz.

medium, the dramatic consequences of the exclusion of volume principle, for a typical non-equilibrium GM system. It is important to remark that below this critical point the GM behaves as a crystal where the particles move in “thermally” activated harmonic oscillations, thus in this regime a Brownian description is suitable, and this fact was already reported by [7]. In a previous work [6] we have studied the spectrum of vibrated GM under gravity, and shown that in the weakly excited regime the dynamics of these evaporated particles precludes describing them as Brownian. They show (over 2 decades) a power law spectrum $S_z(f) \sim 1/f^\nu$ with an anomalous exponent ν ($2 \leq \nu < 5$) that depends on the excited collective movement of the grains. This fact leads us to the conclusion that a description of the present chaotic cooperative dissipative dynamics of the GM particles, must be done in terms of non-Markov particles representing the collective effects of the excited GM [8, 9]. Recent reports concerning the non-Brownian behavior of the velocity fluctuations can also be found in resistivity experiments which show that the electric noise has interesting properties of scale invariance and of intermittency which arise from thermal expansion locally creating or destroying electrical contacts [10]. In addition, associated anomalous transport properties have been reported as non-Brownian motion [11].

Following Hayakawa and Hong [12] we have introduced an approach to understand the behavior of a weakly excited GM [1]. Our 3–dimension N –particles experimental conditions allow us to consider a system of n –rows in a cylindrical container as a 1 dimensional degenerate Fermi-like system. Our laser facility was set up to investigate the occupation dynamics at the fluidized gap that appears at the top of the n –rows GM bed. In Fig. 1(c) a typical $z(t)$ is shown. This measurement corresponds to the beginning of evaporation, where the spectrum exponent is $\nu = 3$, thus we shown that the velocity fluctuation does not correspond to the one that could be obtained from a Brownian particle [6].

By focusing on the configurational properties of an excluded volume theory the s.s. mass profile can be understood in terms of a configurational maximum principle assumption. Excluded volume interactions of the GM do not allow two grains to occupy the same state of gravitational energy. Following Landau’s approach to study a non-equilibrium system, the maximization of $S = \ln \prod_i [\Omega! / N_i! (\Omega - N_i)!]$ yields that the density profile is $\phi(\epsilon) = [1 + Q \exp(\beta\epsilon)]^{-1} \Omega / mgD$ where Ω is the degeneracy (number of boxes in which to put the maximum amount of balls in a constant gravitational layer); β is a Lagrange multiplier parameter, $Q^{-1} = \exp(\beta\mu)$ the fugacity, and $\epsilon = mgDs$ with $s = 0, 1, 2, 3 \dots$. Introducing the normalization we get the relation between the chemical potential μ and the temperature β^{-1} , i.e., $\exp(N\beta mgD / \Omega) = 1 + \exp(\beta\mu)$. The zero-point “chemical potential” is $\mu_0 = mgDN / \Omega$, so N / Ω is the number of

balls in an elementary *column* of diameter D . It is trivial to understand that without pumping of energy the c.m. is characterized by $z_{c.m.} = \mu_0/2mg \equiv h/2$. For a weakly vibrated GM we have shown that β is a non-trivial function of the σ_V^2 [1] (if the particles were Brownons the relation should be $|\bar{v}| \propto \beta^{-1/2}$).

Shaking gently a box of GM is an example of a nonequilibrium system that can be characterized by a global dissipative parameter δ , which in fact is given as the ratio of the two important energies of the system,

$$\delta = 2 \frac{mg\Delta z_{c.m.}}{\mu_0} \left/ \left(\frac{A\omega}{\sqrt{gD}} \right)^2 \right. . \quad (1)$$

Since the motion of these mFp can be studied in terms of the cumulative probability $\phi(\epsilon = mgz)/N$, $z = Ds$ we have shown that at low temperature, $\beta\mu_0 \gg 1$, we get [1]

$$\sigma_z \simeq 2\Theta/mg\beta. \quad (2)$$

where $\Theta = \ln(1 + \sqrt{1 - 1/\sqrt{e}}) + \frac{1}{4}$. This expression gives the amplitude dispersion $\sigma_z = \sqrt{\langle z(t)^2 \rangle - \langle z(t) \rangle^2} = \sigma_\epsilon/mg$ as a function of β . The connection between the fluctuation in the velocity $\sigma_V^2 = \langle V(t)^2 \rangle - \langle V(t) \rangle^2$ and β is given by our generalized equipartition law: $\frac{1}{2}m^*\sigma_V^2 = \frac{1}{2}\frac{\Delta N}{N}\beta^{-1}$, where $m^*/m = \delta$ and $\Delta N/N$ is a relative factor that counts the “active” mFp in the fluidized gap. From this we get

$$\frac{1}{\beta} \left(\frac{1}{\beta\mu_0} \ln [2e^{\beta\mu_0} - 1] - 1 \right) = \delta m \sigma_V^2, \quad (3)$$

and this formula connects σ_V^2 , the dissipative parameter δ and β . So using (1) we relate the collective response of the system: the expansion of the c.m. $\Delta z_{c.m.} = U/N - \mu_0/2$, where $U = \int_0^\infty \epsilon \phi(\epsilon) d\epsilon$, with the variance σ_V^2 and f_e .

Our experiments show that the fluidized gap reveals an inelastic gas heated in a non-uniform way. Now let us understand this gas of mFp by presenting an effective kinetic theory. The collision frequency between mFp is given by $\tau^{-1} = \bar{V}\sigma_0 n$, where $\sigma_0 = \pi D^2$ is the total scattering cross section between two hard spheres, \bar{V} their mean relative speed, and n is the mean number of these mFp per unit volume. The *mean free path* is $L = \tau\bar{v}$. Proposing the relative speed $\bar{V} = c_1\sigma_V$ and the mean speed $\bar{v} = c_2\sigma_V$ (actually \bar{V} should be somewhat larger than \bar{v}) and using that the mean number of “active” mFp per unit of volume is given by

$$n = \frac{4c_3\Delta N}{\pi D_c^2 2\sigma_z}, \quad (4)$$

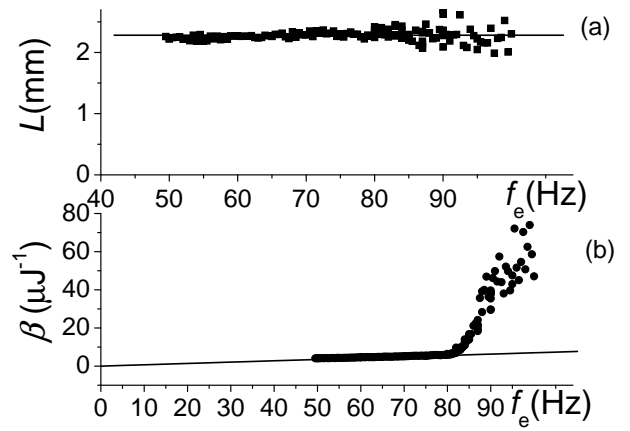


FIG. 3: (a). Mean-free-path L , and (b) the Lagrange parameter β vs. frequency f_e .

and assuming all the constants $c_i = \mathcal{O}(1)$ we get for $\beta\mu_0 \gg 1$

$$L \sim \frac{1}{2} \left(\frac{D_c}{D} \right)^2 \frac{\sigma_z}{\Delta N}, \quad (5)$$

L calculated from the experimental data $\{\sigma_z, \Delta N(\sigma_V)\}$ is depicted in Fig. 3(a) as a function of f_e , in addition in Fig. 3(b) we show β against f_e to note the evaporation transition point. From the linear fitting of Fig. 3(a) we obtain $L \sim 2.28\text{mm}$, this value is indeed only an estimation since we don't know the values of the constants c_i .

With a video camera microscope we took photographs of the surface of the GM-bed from the top of the container and we found that time average pictures show that $L \sim D$, this result agrees with the measurements of correlation in the gas phase [13]. By estimating the degeneration Ω in the expression (5) from $\pi(D_c/2)^2 \sim \Omega D^2$, we get $L \sim 4\Theta D/\pi \ln 2 \sim 1.35D$. This important result shows that a “jumping model” can therefore be considered to build up a kinetic transport theory; thus allowing to calculate the VDF for the mFp in the fluidized gap. We want to emphasize that Fig. 3(a) supports the application of a *jumping model* to study the kinetics of this non-Maxwellian gas because $L \sim D$, i.e., we shall associate a jump probability distribution $P(r)$ (proportional to $\psi(\mu_0 + \epsilon) \left| \frac{d\epsilon}{dr} \right|$, where $r = \epsilon/mg > 0$, [1]) with the transport mechanism of a set of independent particles in a background of a well defined global temperature β^{-1} . A crucial ingredient for understanding the dynamics of the GM would be to be able to build up a statistical mechanics for the construction of the VDF of the driven GM system. A less ambitious program is to have a model for the kinetic transport of the mFp and that is what we are going to present.

Supposing that during the time between collisions the velocity V of each particle is essentially constant, a jump

of length r is performed with velocity $V = r/\tau > 0$; then defining the kinetic energy as $K = mV^2/2$ and using the transformation of random variables we get $P(K)dK = P(r)dr$. This “jumping model” allows us to arrive at a closed expression for the density of the kinetic energy in terms of the β of the s.s. of the configurational entropy, i.e.,

$$P(K) \propto g\tau \sqrt{\frac{m}{2K}} \psi(\mu_0 + g\tau \sqrt{2mK}); \int_0^\infty P(K) dK = 1. \quad (6)$$

Using the mass profile we get $\psi(\epsilon) = -d\phi/d\epsilon$, therefore from (6) we obtain an analytic formula for the kinetic distribution function (KDF) $P(K)$, which predicts at medium and high energy ranges a larger population than the Maxwellian. Note that in the asymptotic behavior $\epsilon \gg \mu_0$, $\psi(\epsilon) \rightarrow \beta e^{-\beta\epsilon}$ then KDF for high energy goes like $\sim g\tau \sqrt{m/K} e^{-g\tau\beta\sqrt{2mK}}$ which is non-Maxwellian.

An important program is to perform a suitable experiment to be able to determine the functional form of the KDF, and this is what we are going to present in the rest of the paper. While in the first experiment we scan in f_e at a given Γ to investigate the behavior of β , in a second experiment we carry out an isothermal-like measurement during a long enough time to get the KDF and the corresponding β .

From the measurement of $z(t)$ we were able to calculate the time dependence of the kinetic energy flux in the measurement of a 1-dimensional window, from which it is possible to obtain the KDF. At a given f_e and Γ we measure 5000 windows of 2 seconds to get a total of 25 millions of events from which we build up the s.s. KDF. In Fig. 4 we fit our experimental data by using the formulae (6); the experiment was carried out for fixed $f_e = 80\text{Hz}$ and $\Gamma = 10$. Medium and high energy *particles*, which are at the surface of the vibrated GM bed, are well described by formulae (6), while at low energy the behavior is Maxwellian. Also we fit the whole experimental results with a Maxwellian distribution in kinetic energy $\Pi(K) = \exp(-K\beta)/\sqrt{\pi K/\beta}$. As well as with a stretched exponential for the velocity distribution: $\Pi(V) \sim \exp[-(V/V_0)^\alpha]$, with $\alpha = 3/2$, where V_0 is the thermal *rms* velocity[14]; this last fitting shows that the agreement is also good. Since Olafsen et al. reported the stretched exponential law for the VDF of the gas phase of a vibrated monolayer GM[13], several research groups have been measuring this law for granular gases [15]. A delicate obstacle for the analytical solution of this problem is the supply of energy to the granular bed which *compensates* the dissipation caused by inelastic collisions [14], an aspect which was clarified by van Zon et al. using simulations of molecular dynamics [16]. For granular gases there is indeed a family of distributions, with apparent exponents $\alpha < 2$ which are governed by the ratio between the numbers of heating events and in-

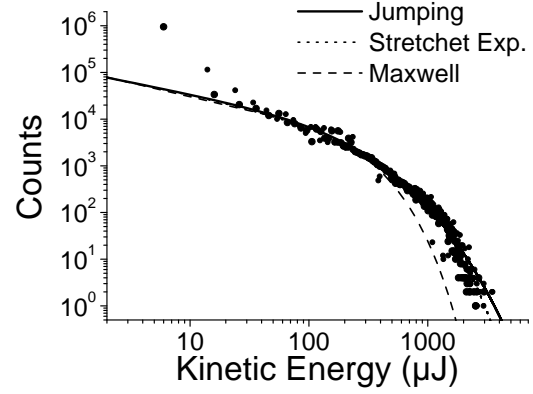


FIG. 4: Experimental kinetic energy distribution function (KDF) and the corresponding fittings according to our “jumping” kinetic model, the Maxwellian and the stretched exponential cases.

elastic collisions [16]. Either under uniform heating or boundary heating of the GM, if the dissipative collisions dominate the heating process produces a liquid-like cluster surrounded by a gas phase and the VDF is strongly non-Gaussian. Also after Olafsen et al. the conclusions of several authors keep clear the conceptual relationship between this high energy tail behavior, the exclusion of volume and the clustering in granular gases. Our description adds a new picture for clarifying this delicate point, the δ parameter defined in equation (1) and the generalization of the equipartition law (3) permit us to tackle this obstacle, to give the KDF in terms of β . Our approach was obtained from the physical parameters: $\{f_e, \Gamma, \mu_0, D, N, \Omega\}$ and by using the free parameter δ from the experiment described in Fig. 2 and 3. The fitting for the curve in Fig. 4 gives $\beta = (1.7 \pm 0.5)(\mu\text{J})^{-1}$ and for the corresponding value from Fig. 3(b) we get $(5 \pm 1)(\mu\text{J})^{-1}$, which in fact is a good agreement considering that in the first experiment we are scanning in f_e at constant rate, and in the second experiment (time average) both f_e and Γ are fixed. The high energy tail corresponds to the beads near to the surface of the GM bed where the jumping combined with collisions dominates the process.

A point of special interest is that the *packing fraction* for this fluidized gap is constant for the whole range of low temperatures we used, which can be estimated as:

$$p_f = \frac{2}{3} \left(\frac{D}{D_c} \right)^2 \frac{D \cdot \Delta N}{\sigma_z} \sim \frac{\pi \ln 2}{12 \Theta} = 0.25. \quad (7)$$

At constant cooling down by increasing f_e at constant Γ , this lattice-gas condenses into an *fcc* crystal, and vice-versa from the *fcc* lattice under constant heating rate the expansion of the c.m. entails the expansion of this *fcc*. This expansion under gravity creates fluctuations in the number of particles $n(\epsilon)$ around $\epsilon = \mu_0$, which induces

the transport of matter at the fluidized-gap. The time integration of such fluctuations (as we did during 3hs. in [1]) reveals that the fluidized-gap has a structure with a Fermi profile. Thus we conclude that for our experimental conditions the gaseous gap is indeed a lattice-gas (*fcc* where half of sites are occupied) with p_f according to equation (7). Nevertheless, the picture we get for this lattice-gas should be understood as a “jammed state of motion”, since the p_f does not change for the range of low temperatures we measured.

The important experiment of D’Anna et al. [7] shows that a vibrated granular bed exhibits a formal analogy with a *thermal bath*, where it is possible to apply a fluctuation-dissipation theorem. In that experiment a harmonic oscillator of frequency ω_0 (the torsion pendulum) is coupled to the Brownian particles in the vibrated granular bed. This thermal bath is generated by driving the GM in a frequency band between 300 – 900Hz at a given Γ (from 1 to 11.6g); in fact, out of equilibrium this thermal bath is thought within the frame in [2]. The D’Anna’s torsion pendulum is this thermometer coupled to the Brownian ensemble, the driven fluctuation pumps energy while its interaction with the same Brownons provokes the dissipation of its energy. Nevertheless if evaporation starts the viscosity of the thermal-like bath should therefore be related to the exclusion of volume, since the jumping of the particles is conditioned by the availability of free volume.

In a system in equilibrium and in D’Anna’s experiment, equipartition means that K must be $k_B T/2$ and by measuring long enough they get a temperature for the thermal bath (or granular bed), the range of temperatures tested were lower than $0.1\mu\text{J}$. Our laser-thermometer explores beyond the Markovian range of the GM bed, which happens when the jumping events start (evaporation) and the fluidized gap appears. Our thermometer is a test particle of the GM bed which occupies the position tested by the laser.

Measurement of β^{-1} as a function of f_e exhibits at 10 PKelvin the critical point that characterizes the evaporation transition. For $\beta^{-1} < 10\text{PKelvin}$ ($\beta^{-1} < 0.1\mu\text{J}$) the system is condensed and the motion corresponds to Brownian oscillators. If evaporation starts the following picture is possible: when our ensemble of mFp is perturbed, the energy is dissipated by the mechanisms of jumping (under gravity) and random inelastic collisions between the mFp , but conditioned to the fact that these events are self-organized in time, keeping constant the p_f of the fluidized gap. The relation between the transport and the number of “active” particles ΔN (at the Fermi sea) links the two Lagrange parameters: β and μ (compactivity), and therefore describes the existence of a lattice-gas. The coupling between β^{-1} and μ tells us that the important rule, for the ensemble of mFp , is to dissipate energy keeping constant the lattice structure. We remark that the difference with the well known

Markovian motion is that Brownons can dissipate energy through viscosity independently of any gas structure.

Our approach focuses on the potential energy which is a function of the configurational entropy and the relation with the kinetic energy per active particle at the gap. In other words, while in the Brownian situation we can describe the system in terms of energy fluctuations, viscosity and temperature, beyond the Markovian approximation, the important events are the fluctuations in the number of particles. Then we are indeed reporting on the existence of a *compensation* relation between the fluctuations in the number of mFp at the gap (or the matter transport), the Fermi profile held by the δ dissipative parameter, and the global temperature β^{-1} . A fluctuation in the number of mFp in the gap will be compensated in time in such a way that the lattice-gas keeps p_f constant: this is a fluctuation-compensation relation. Therefore a stochastic formalism is still valid but the equations should describe a fluid where the complexity is the description of the lattice-gas, the jumping phenomena and collisions in a space framed by the exclusion of volume.

This approach agrees with the discussion in [2] related to the idea that systems far from equilibrium can be described with more than one temperature. Such temperatures are related to time constants of the different configurational structures. For a glass, the time for relaxation to equilibrium is extremely long, and it is possible to describe this system by using more than one thermometer with different time constants. In GM experiments, the structure of the gap is only measurable if we wait enough time. This structure is related to transport of mass at the gap, as is revealed by our generalized equipartition law. The characteristic time we must wait to measure β in this system is the time we need to integrate the mass profile. However unlike glasses, GM vibrated under gravity is far away of equilibrium but stationary: the system exists while the δ parameter is held at a fixed value, and the fluctuation-compensation should play a role stabilizing the steady state, i.e., the precise *balance* of fluctuations permits us to know the global temperature without waiting too long.

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